Acid deposition and its ecological effects: a brief history of research

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Abstract

Research on acid deposition can be traced to the mid-19th century, but has expanded greatly in the last 50 yr, focussing in particular upon the effects on ecosystems of acid deposition caused by the combustion of fossil fuels. Observational studies first associated such acidification with its chemical and biological effects. They were supplemented in 1976 by experimental acidification of a whole lake. By 1980, paleoecological techniques were employed to investigate past histories of acidification. In the 1980s, critical loads were calculated to measure how much acid deposition could be tolerated by aquatic ecosystems. Reductions in emissions of sulfur dioxide, as well as experimental studies in the mid-1980s, allowed investigations of recovery from acidification. At the same time, other studies showed that lake sediments and wetland peats mitigate acid deposition by reducing sulfate and nitrate. In the mid-1990s it has become apparent that acid deposition interacts with other environmental stresses such as climate warming and ozone depletion.

Keywords: Acidification; Biogeochemistry; Critical load; Forest; Interaction; Lake; Neutralization; Policy; Recovery; Wetland

1. Introduction

This paper presents a brief review of major advances in the history of research on acid deposition and its effects on lakes, forests and wetlands. It deals with topics such as the sources of acidifying and neutralizing materials in the atmosphere, the acidification of ecosystems, factors involved in the resistance of ecosystems to acidification, the definition of critical loads for ecosystem protection, and effects of acid deposition on organisms. Other topics considered include natural causes of ecosystem acidification, recovery from acidification, interactions of acid deposition with other environmental stresses, the methods used to study acid deposition and its effects, major international meetings, and policy lessons. A chronology of major advances in research, somewhat idiosyncratic because it has been compiled by a single author, is also presented.

An attempt has been made, for each advance mentioned in the text, to cite the paper in which the discovery was first reported. (Inevitably I must have failed in some cases to identify the earliest report, for which I apologize.) The cited studies have not been evaluated in depth, nor have I attempted a full review of where each of them has led the science associated with acid deposition. Furthermore, I have only touched briefly on major controversies associated with that science.

2. Early history

Our scientific knowledge of ecosystem acidification goes back at least to the mid-18th century, when Home (1757) (see also Gorham, 1989) applied the concept of acids, bases and neutral salts to soils. Acid deposition was described a century later, when Ducros (1845), using the term 'pluie acide' (acid rain), analyzed hail from a thunderstorm in France and found it to be acidified by nitric acid formed naturally by electric discharges during the storm (an editorial comment by E. Frémy stated that Ducros confirmed a fact long known to science). It is worth noting that Ducros was careful to test for, and exclude, carbonic, hydrochloric and sulfuric acid as possible alternatives. A few years
later Smith (1852) analyzed rains in and around the city of Manchester in England, noting that in the city itself they were acidified by sulfuric acid produced as a consequence of coal combustion. Twenty years later he published a book, ‘Air and rain: the beginnings of a chemical climatology’ (Smith, 1872), that laid out many of the basic ideas concerning acid deposition (Gorham, 1981, 1982). After the turn of the century Crowther and his colleagues (Crowther and Ruston, 1911; Crowther and Steuart, 1913; see also Cohen and Ruston, 1912) described a gradient of increasing precipitation acidity toward the center of the city of Leeds in England, and demonstrated that both acid rain and dilute sulfuric acid had detrimental effects on the growth of plants, the width of tree rings, seed germination, microbes involved in the nitrogen cycle, and soil quality. They ascribed the acidity of Leeds rain to sulfuric acid, although they noted that both sulfate and chloride increased toward the city center (British coals were rich in both elements). In fact, analysis of their data by partial correlation (Gorham, 1981) revealed that acidity correlated with chloride rather than sulfate (see also Gorham, 1958a).

3. Acidification and neutralization

The modern era of investigations concerned with acid deposition from the atmosphere began in the mid-1950s, although scattered observations of low pH in rain were made before that time (Gorham, 1981). Cauer (1949) had also observed acid fog-condensation nuclei in Germany, some of which, near industrial areas, he ascribed to sulfuric acid from pollution, and some, in more remote mountain areas, to very dilute nitric acid formed naturally. It was in 1955 that a distinct correlation between hydrogen ions and sulfate ions was observed regionally by Barrett and Brodin in precipitation in rural Scandinavia, and locally by Gorham (see also Gorham, 1958b) in precipitation in the rural English Lake District. At the same time Houghton (1955) reported a similar correlation in fog and cloud water from the mountains of New England. These observations demonstrated that acid deposition caused by fossil-fuel combustion was subject to long-range transport from urban centers of air pollution. In the 1960s acid deposition caused by metal smelters in Ontario was inferred from the decline of both hydrogen ions and sulfate ions in pond and lake waters at varying distances away the sources of sulfur-dioxide emissions (Gordon and Gorham, 1963; Gorham and Gordon, 1960).

Reviews of the early history of research on acid deposition have been written by Gorham (1981) and by Cowling (1982); see also a more general review of atmospheric deposition by Gorham (1992). The history of acidification research in Sweden has been described by Tamm (1995).

Studies of precipitation chemistry in remote areas (Galloway et al., 1982) indicated some acidity owing both to simple organic acids (formic and acetic) and to sulfuric acid. When contributions from human activities were removed, the natural volume-weighted mean pH of precipitation in these remote areas appeared to be 5 or above.

Neutralization of acid deposition was also considered in the early studies. Obvious sources were bases from fly ash produced by coal combustion, or from blown soil dust (Gorham, 1955, 1957, 1958b; see also Fisher, 1968). Cement dust could be important locally (Barrett and Brodin, 1955). Ammonia from agricultural sources was implicated by Barrett and Brodin (1955); it reacts with sulfur dioxide and sulfuric acid to form ammonium sulfate in the atmosphere (Junge, 1954). Gorham (1976) provided an early review of the subject.

It is noteworthy that acid precipitation contains a variety of trace metals, as was demonstrated in Japan by Morita (1955), and in the 1970s by a number of other investigators (Gorham, 1976). The contrast between the influence of acid deposition and the deposition of particulates from prairie soils was shown clearly in the early 1980s along an east–west transect of precipitation samples in Minnesota, in which both major and minor elements were analyzed (Eisenreich et al., 1980; Thornton et al., 1981). The presence of organic micropollutants in Norwegian precipitation was reported by Lunde et al. (1977).

4. Diverse causes of acid precipitation

Although sulfuric acid has been the primary cause of low pH in precipitation, the local importance of hydrochloric acid in cities of northern Britain has been mentioned. The importance of nitric acid from fossil-fuel combustion, especially in automobiles, was indicated by Likens et al. (1972). A further cause of ecosystem acidification is ammonium sulfate, which is often abundant in precipitation. Although the reaction of ammonia with sulfur dioxide or sulfuric acid in the atmosphere produces a neutral salt, it has a strong acidifying effect upon soils, as has long been known by agricultural scientists (Gorham, 1976, 1989). The great importance for ecosystem acidification of the deposition of ammonia and its reaction product with sulfuric acid has been emphasized recently by Galloway (1995), who has pointed out that by the year 2020 Asian emissions of sulfur dioxide, oxides of nitrogen, and ammonia will equal or exceed those of Europe and North America combined. Postek et al. (1995) recently demonstrated experimentally the significance
of ammonium sulfate in acidifying streamwater and enriching it in inorganic aluminum.

All of these acidifying and neutralizing substances may be deposited by rain-out within clouds or wash-out below cloud base. Alternatively, they may occur as dry particulates that fall out of the atmosphere (Cohen and Ruston, 1912) or are impacted on surfaces such as tree leaves and needles (Eriksson, 1960; Gorham, 1976). Fowler (1980) provided an early review of both wet and dry deposition of sulfur and nitrogen compounds from the atmosphere (see also Fowler et al., 1991).

5. Effects on ecosystem biogeochemistry

Acid deposition has been shown to affect the chemistry of lakes, forests and wetlands in a variety of direct and indirect ways. Only since the 1980s, however, have statistically reliable estimates of its regional effects on ecosystems been gathered, for lakes in the United States (Linthurst et al., 1986).

5.1. Lakes

The effect of acid deposition on the pH of lakes was recognized at the same time that it was observed as a phenomenon in the rural English Lake District. Gorham (1955, 1958a,b) and his colleague Mackereth (1957) reported that small lakes in the central mountains of the district, lying on hard rocks that release bases very slowly, were being strongly acidified by sulfuric acid, in contrast to lakes on softer, more easily weathered rocks around the periphery of the district, whose ions were strongly dominated by calcium bicarbonate. Soon afterward it was noted (Gordon and Gorham, 1963) that acid deposition close to the iron-smelting plant at Wawa, Ontario, was causing substantial leaching of calcium from local soils into adjacent ponds and lakes. Much later it was observed by Wright et al. (1976), and by Cronan and Schofield (1979), that acid deposition led to dissolution of aluminum in soil minerals and to a strong enrichment of aluminum ions in lake water.

Effler et al. (1985) showed that concentrations of dissolved organic matter were lowered in lakes acidified by acid deposition, owing primarily to coagulation and sedimentation of dissolved humic compounds by aluminum or to changes in the properties of dissolved organic matter. This finding was confirmed in experimentally acidified lakes by Schindler et al. (1992). An associated physical result of acidification was greatly increased transparency, observed earlier by Almer et al. (1974) and Yan (1983).

An alternative hypothesis that lake acidification has been caused much less by acid deposition than by changes in land use, notably regrowth of forests after cutting, was proposed by Rosenqvist (1978) in Norway Rosenqvist (1978) and by Krug and Frink (1983) in the United States. The hypothesis has not stood the test of time, as pointed out in a review by Schindler (1988). Slow natural acidification of lakes because of the leaching of bases from catchment soils has been demonstrated, most clearly by Renberg (1990).

As already noted, fossil-fuel combustion results in emission of trace metals such as lead and zinc, relatively high concentrations of which were observed in southern Norwegian lakes subjected to acid deposition (Henriksen and Wright, 1978). Galloway and Likens (1979) observed that trace metals were enhanced for a similar reason in the uppermost sediments of lakes in New York State. Trace metals had earlier been found to be substantially enriched in lakes subject to smelter pollution (Stokes et al., 1973), partly owing to emissions from the smelters (Gorham and Gordon, 1963; Hutchinson and Whitby, 1974) and presumably also to acid leaching of mine spoil.

Deposition of both ammonium and nitrate ions along with sulfate has provided a certain degree of nitrogen enrichment in lakes. It is, however, far less significant than other causes of cultural eutrophication.

5.2. Forests

The first clear evidence of acid deposition upon the acidity of forest soils was that of Rusnov (1919) in Austria, who ascribed the effect to sulfur dioxide from air pollution. Twenty years later Katz (1939) demonstrated that soils around a lead–zinc smelter in British Columbia were acidified by emissions of sulfur with a consequent loss of base saturation. More recently Hallbäcken and Tamm (1986) described decreases in the pH of forest soils (independent of stand age) in Sweden between 1927 and 1982–1984, owing in part to acid deposition from distant sources. Very recently Likens et al. (1996) reported the loss over three decades of large quantities of calcium and magnesium from soils in the Hubbard Brook Experimental Forest in New Hampshire. They ascribed the cause to both leaching by acid deposition and a concurrent decline in the atmospheric deposition of base cations (Hedin et al., 1994). Another indirect result of the influence of acid deposition on soils is the mobilization of soluble aluminum ions from soil minerals (Ulrich et al., 1980).

A direct result of acid deposition on forest soils is their enrichment in both ammonia and nitrate, much of which is usually retained and stored in biomass. In severe cases, however, nitrate saturation of the ecosystem can occur, so that atmospherically derived nitrate passes directly through the soil and into streams and lakes (Grennfelt and Hultberg, 1986). In the lakes of
southern Norway, for instance, nitrate concentrations approximately doubled over the 12 years from 1974–1986 while atmospheric deposition of nitrogen increased only slightly (Henriksen et al., 1988).

5.3. Wetlands

The further acidification of already acid peat bogs by acid deposition was reported by Gorham (1958b), and more recently by Vangenechten (1980). The base saturation of peats has also declined (Skiba et al., 1993). Circumneutral fens are expected to be sensitive to acid deposition (Gorham et al., 1984a, 1987). Those anticipated to be most sensitive have surface-water alkalinities less than 40 meq l\(^{-1}\) and a pH of about 6.

Acid deposition is, however, mitigated by microbial reduction processes, as reported by Hemond (1980) and by Gorham et al. (1985).

Gorham (1958b) suggested that contamination by trace metals as well as by acid deposition was likely, and Hvatum (1972; cited by Cowling, 1982) showed an increase in lead concentration near the surface of Norwegian peat bogs that he interpreted as a result of the long-distance transport of air pollution. Radioactive fallout, for instance, \(^{137}\text{Cs}\) and \(^{239}\text{Pu}\), also tends to be concentrated in the surfaces of peat deposits (Oldfield et al., 1979), along with organic micropollutants (Rapaport and Eisenreich, 1986).

A quite different aspect of influence upon peatlands is the atmospheric deposition of nitrogen upon them. Whether this is leading to increased sequestration of nitrogen in peat appears as yet unknown.

5.4. Proton budgets

The utility of proton budgets as a way of accounting for sources and sinks of acidity was demonstrated in forest soil subject to acid deposition by Mayer and Ulrich (1976) (see also Ulrich, 1980). A similar study of a peat bog, to which the sole source of input was atmospheric deposition, was carried out by Hemond (1980) in Massachusetts. An interesting comparison of the Massachusetts bog with one in Minnesota not subject to significant acid deposition, but instead to considerable dustfall from soil, was made by Urban et al. (1987) a few years later.

6. Factors involved in the resistance of ecosystems to acidification

There are many factors that confer resistance to acidification (otherwise known as acid neutralizing capacity) upon ecosystems. First among them is the base-cation content and weatherability of rocks (Gorham, 1958b; Mackereth, 1957) and the associated degree of base saturation in the soils derived from them, or in the peats associated with them (Gorham et al., 1984a). A process particularly important in old, highly leached and strongly weathered soils is the adsorption of sulfate on oxides of iron and aluminum (Khanna and Beece, 1978; Wikander, 1975), which retards leaching of soil bases as sulfates. The thickness of soils and the slope of the land surface, which govern how effectively atmospheric precipitation percolates into the soil or runs off over and through the leached surface soil horizon, are also important, particularly in situations where an accumulation of acids in the snowpack in cold climates can result in a sudden acid pulse when the snow melts (Galloway et al., 1980).

A significant aspect of resistance is the internal generation of alkalinity by reduction of sulfate. The phenomenon has been known since the 1930s, but was first recognized in the context of lake acidification by Schindler et al. (1980) and of bog acidification by Hemond (1980). According to Norton et al. (1988), however, alkalinity generation in drainage lakes is usually dominated by terrestrial processes, whose influence declines as the ratio of catchment area to lake area decreases. They found that in the northeastern United States, where acid deposition is relatively severe, sulfate reduction accounted for less than 10% of the non-marine sulfate present in lake waters. Cation exchange, between hydrogen ions in the water and calcium and magnesium ions adsorbed on the cation-exchange complex of the uppermost sediment, is also involved in mitigating lake acidification (Cook et al., 1986; Schindler et al., 1991).

7. Critical loads

Once it was recognized that acid deposition was causing damage to aquatic ecosystems (chiefly by sulfuric acid, because nitric acid was utilized by the biota), efforts began to determine a critical load for sulfate below which such damage would not occur. (The term ‘target load’ can be used for a less rigorous, and more expedient, first objective.) Early efforts focussed on estimating the load that would result in an annual average pH in precipitation of 4.6–4.7, because in areas below that range damage was observed in sensitive aquatic ecosystems (Anonymous, 1981; Henriksen, 1979). Canadian members of work group I on impact assessment, in their 1983 final report under the US-Canada memorandum of intent on transboundary air pollution, indicated a target wet-sulfate load of 20 kg ha\(^{-1}\) yr\(^{-1}\) as appropriate. Shortly thereafter Gorham et al. (1984b) suggested that this was too high for eastern North America, and proposed a lower limit of 14–16 kg ha\(^{-1}\) yr\(^{-1}\), which was still
well above the estimated natural load of about 10 kg ha\(^{-1}\) yr\(^{-1}\).

In the mid-1980s Scandinavian scientists (Nilsson and Grennfelt, 1988) took another approach, calculating the critical loads of sulfate that would produce a surface-water pH of 5.3–5.5 in terrains of different soil type. These loads ranged from as little as 7.2 kg ha\(^{-1}\) yr\(^{-1}\) on the most sensitive, shallow organic soils with low calcium and magnesium contents to as much as 19 kg ha\(^{-1}\) yr\(^{-1}\) on highly weathered silty clay loams. At the same time they applied the concept to forest ecosystems. Critical loads for surface waters have been related to acid neutralization capacity by Henriksen et al. (1995). Maximum ‘allowable’ annual deposits of hydrogen ions for five classes of European ecosystems have been defined by Chadwick and Kuylenstierna (1991) (cited by Last, 1991). These range from 20 kg H\(^+\) km\(^{-2}\) yr\(^{-1}\) for their most sensitive ecosystems to >160 kg H\(^+\) km\(^{-2}\) yr\(^{-1}\) for the least sensitive class.

Critical loads for nitrogen in waters and soils were discussed in a report edited by Nilsson and Grennfelt (1988). Very recently, Bobbink and Roelofs (1995) have estimated critical loads of nitrogen to a variety of ecosystems (softwater lakes, fens, ombrotrophic bogs, grasslands, heathlands, and coniferous and deciduous forests). For the first two (the most sensitive ecosystems) they proposed a critical load of 5–10 kg ha\(^{-1}\) yr\(^{-1}\); the average for all ecosystems they studied was 15–20 kg ha\(^{-1}\) yr\(^{-1}\).

8. Effects on organisms

Acid deposition affects, both directly and indirectly, a wide variety of organisms in a multitude of different ways. Some examples follow.

8.1. Aquatic biota

In 1959 Dannevig recognized that where Norwegian surface waters were acidified by atmospheric deposition, fish tended to disappear (see also Jensen and Snekvik, 1972). An early indication of the effect of acid deposition upon aquatic plants was provided by Gorham and Gordon (1963), who showed a marked decline in the diversity of aquatic macrophytes in ponds and lakes close to the Sudbury smelters. Whether the effect was due mainly to acidity or to the toxicity of associated copper and nickel was uncertain; probably both were involved; Stokes et al. (1973) were able to demonstrate the development of tolerance to heavy metals in algae isolated from lakes polluted by the Sudbury smelters. By the end of the 1970s many other effects had been observed (Drablos and Tollan, 1980), including changes in communities of aquatic plants and animals and a lessening of their diversity below pH 6.0.

Effects upon fish were investigated in considerable detail and included inhibition of reproduction (Beamish and Harvey, 1972), loss of salt balance (Leivestad and Muniz, 1976), accumulation of mercury (Almer et al., 1978), and toxicity of aluminum (Cronan and Schofield, 1979). Havas et al. (1995) have provided a recent overview. In the 1980s loss of lake trout owing to elimination of several organisms in the food chain was demonstrated by Schindler et al. (1985) in an experimentally acidified lake. Acidification can favor parasitism and impair reproduction, as shown by France (1982, 1987) in a study of crayfish in the same lake. Muniz (1991) and Schindler et al. (1991) have provided detailed accounts of the biological effects of acidification in lakes. Comparison of atmospherically and experimentally acidified systems has indicated substantial agreement in the responses of both biogeochemical processes and effects on the biota of lower trophic levels, which generally declined in diversity.

8.2. Forest trees

In terrestrial ecosystems the chief emphasis has been on a possible linkage between acid deposition and forest decline (Ulrich et al., 1980). Studies began in Europe in the early 1970s (Overrein et al., 1980), and accelerated greatly in the 1980’s (Schulze and Freer-Smith, 1991). Pietelka and Raynal (1989) reviewed the general topic, and provided a follow-up discussion of the role of science and scientists in dealing with the problems. That discussion was supported by Stout (1989) but criticized (in my view appropriately) by Woodwell (1989). Numerous effects have been postulated, including loss of soil bases, aluminum toxicity, interactions with air pollutants such as ozone and sulfur dioxide, and interaction with natural phenomena such as drought and deficiencies of soil nutrients. In North America, studies in the 1980s and 1990s indicated that acid cloudwater reduces the resistance of montane red spruce to freezing injury (Johnson et al., 1996) and that heavy metals may also be present in toxic concentrations (Gawel et al., 1996). Saturation of the soil solution by atmospheric deposition of nitric acid, which can leach base-cation nutrients from soils, has also become an issue of serious concern (Grennfelt and Hultberg, 1986; Raloff, 1995). It appears to act in part through diminution of the biomass of fine roots (Matzner and Murach, 1995). The entire situation is obviously extremely complex, and has yet to be elucidated satisfactorily. Experimental additions of nitrogen to forest ecosystems should be especially informative in this connection, and Tietema et al. (1995) reviewed several such experiments devised in the 1980s in the context of acid deposition to forests.
Nitrogen is well known to limit the productivity of many ecosystems, which may respond by increased productivity but also by marked changes in species composition and losses of biodiversity. The long history of research on nitrogen fertilization of terrestrial ecosystems (for an excellent early example see Mitchell and Chandler, 1939) has been reviewed by Tamm (1991).

8.3. Bog mosses

Ferguson et al. (1978) showed that the growth of Sphagnum species is susceptible to both fumigation by sulfur dioxide and treatment by sulfuric acid at concentrations comparable to those in northern Britain, where these mosses have undergone a major decline over the past century and a half (Tallis, 1964). Nitrogen metabolism may also be altered by inputs of ammonia and nitrate in acid deposition (Press and Lee, 1982; Woodin et al., 1985).

8.4. Modification of the effects of acidity on organisms

It has long been known that the distribution of various organisms in relation to habitat pH can be modified by other variables. An especially good set of examples has been provided by a recent study of widely different animals (gastropods, two species of fish, Daphnia, and loon broods) in the lakes of southeastern Canada (Doka et al., 1997). The probability of their occurrence, lower at higher acidities in all cases, has been plotted versus pH for three specified levels of other variables. In the case of gastropods, which occur frequently at circumneutral pH levels but only rarely below pH 5.0, it has been shown that higher concentrations of aluminum and sulfate ions decrease their probability of occurrence at any given level of pH. With increasing acidity northern pike show a decline in their probability of occurrence, which is, however, much less the larger the lake and the cooler the temperature. Pumpkinseed tend to be favored in lakes of much less the larger the lake and the cooler the temperature. Pumpkinseed tend to be favored in lakes of much less the larger the lake and the cooler the temperature.

9. Natural causes of ecosystem acidification

Natural causes of ecosystem acidification have been known for a long time. For instance, Thaer (1810) recognized the role of carbonic acid in chemical weathering and remarked that humus, the residue of organic putrefaction, was slowly converted into carbonic acid by the action of oxygen. The presence of complex ‘humic acids’ in soils was demonstrated soon afterward by Sprengel (1826), and these acids were then associated with leaching and the formation of acidic podzol soils. They were also associated, by Munzt and Marciano (1888), with the acidity observed in tropical blackwater streams. Salisbury (1925) was the first ecologist to describe a dated chronosequence of soil acidification, in English sand dunes. Natural acidification of a Swedish lake over thousands of years was documented in detail by Renberg (1990). The general topic of ecosystem acidification has been reviewed by Gorham (1989).

Acid deposition itself is not caused only by human activities. For instance, Bottini (1939) found rain around the volcano Vesuvius in Italy to contain hydrochloric acid, and Hutchinson et al. (1979) observed ponds with pH values as low as 1.8, and extremely high sulfate concentrations, around the Smoking Hills in arctic Canada, where natural fires in exposed lignite beds generate large amounts of sulfur dioxide.

Ecosystems can be acidified naturally by other means. Yoshimura (1933, 1935) reported biotic impoverishment associated with unusually low pH levels and high sulfate concentrations in volcanic lakes in Japan. Anthropogenic illustrations of the same phenomenon have been provided by acid mine drainage (Baker et al., 1991), by the drainage of swamps rich in sulfides (Chenery, 1953), and by other land disturbance (Kerekes et al., 1986; Ohle, 1935).

A natural type of acidification of great importance in the boreal zone is the export of complex colored organic acids from peat bogs (Gorham et al., 1985; Thompson et al., 1927) into streams and lakes (Gorham et al., 1986; Gorham et al., 1998; Kortelainen, 1993). According to Kortelainen and Saukkonen (1995), colored organic acids dominate stream-water acidity over large parts of Finland outside the south, where acid deposition is important. The same is true of central and southwestern Nova Scotia (Gorham et al., 1998), except around the urban center of Halifax (Gorham et al., 1986). Although such bogs reduce much of the sulfate and nitrate falling upon them (Gorham et al., 1985, 1998; Hemond, 1980), the acids they export act additively with acid deposition falling on upland soils to lower the pH of lakes and streams (Gorham et al., 1986).

Another (relatively minor) cause of surface-water acidification, the so-called sea-salt effect, was described by Wiklander (1975) and later confirmed by many others. Wiklander suggested that stormy episodes of high sea-salt deposition would cause hydrogen ions and aluminum ions to be released into solution owing to their replacement on the cation-exchange complex of the soil by marine base-cations.
10. Recovery from acidification

Investigations of the recovery of acidified aquatic ecosystems in response to both lowering of sulfur emissions and reduction in experimental additions of acid began in the 1980s. Toward the end of the decade a good deal of information about responses to emission reductions had been gathered around Sudbury and in other parts of Ontario, as well as in other areas of North America and in southwest Scotland. Data on experimental reversal of acidification became available as well from both Ontario and Norway. The results of these diverse investigations have been summarized by McNicol et al. (in press); Schindler (1988), Schindler et al. (1991) and Wright and Hauhs (1991).

Once the extent of lake acidification became apparent, liming was used as a treatment to prevent damage and hasten recovery. The practice has been widely used in Sweden since 1977, and by 1985 3000 lakes and 100 streams had been treated (Lessmark and Thörnelöf 1986). Effects on crayfish and fish in selected lakes and streams were reported by Nyberg et al. (1986); crayfish benefited little but fish numbers increased, although catch in weight per unit effort remained about the same.

10.1. Responses of precipitation chemistry to declining emissions

According to McNicol et al. (in press), since 1980 there have been substantial declines in emissions of sulfur both in Canada (53%) and in the United States (20%). Data on sulfate in precipitation generally show declines, accompanied by reductions in acidity that have not, however, been equivalent to the declines in sulfate. An important reason is that substantial declines have occurred simultaneously in the atmospheric deposition of base cations, owing to reductions in particulate emissions from urban and industrial sources and, probably, in dustfall from unpaved roads (Hedin et al., 1994; see also Gorham, 1994). Similar reductions may have occurred as agricultural soils have become subject to no-till cultivation.

10.2. Chemical responses of aquatic ecosystems to lowered acid deposition

In Norway, studies of lake-water chemistry have revealed that reductions in sulfate have been offset by increases in nitrate. Because of this, concentrations of non-marine strong anions and the potential for soil acidification have remained the same (Henriksen et al., 1988).

In Canada, Jeffries (1997) reported that of 202 soft-water lakes in eastern Canada that have been monitored since 1980, pH levels remained about the same in 56%, increased in 33%, and decreased in 11%. In the case of the Sudbury metal smelters Schindler (1988) remarked that a combination of smelter closures and controls on emissions of sulfur reduced those emissions to about a third of the levels observed in the early 1970s. This resulted in decreased acidity of atmospheric deposition and a lowering of sulfate concentrations in nearby lakes, together with a consequent rapid increase in alkalinity and pH. Concentrations of aluminum and trace metals declined at the same time.

At Hubbard Brook in New Hampshire, concentrations of both sulfate and base cations have fallen substantially in stream water in response to similar concentration declines in precipitation (Driscoll et al., 1989). About 80% of the decline in base cations can be explained by decreases in the inputs from precipitation. Because of the concurrent decline in both sulfate and base cations, the range of stream pH has remained about the same, between 4.8 and 5.0.

In the Adirondack mountains of New York state, Driscoll et al. (1995) observed a uniform decline of lake-water sulfate in recent decades, but to a lesser extent than anticipated from the decline in sulfate deposition, perhaps owing to release of previously deposited sulfate from soils. Basic cations in lake waters declined concurrently, while pH and acid-neutralizing capacity showed little change.

At Galloway in Scotland, Wright and Hauhs (1991) cited independent lines of evidence for lake recovery following reductions of about one-third in the concentrations of non-marine sulfate in precipitation. Regional surveys of lake-water chemistry carried out in 1979 and repeated in 1988 revealed substantial declines in non-marine sulfate, the sum of calcium and magnesium, and labile aluminum, indicating less acid weathering. An individual study of Loch Enoch over several years from 1979–1989 showed distinct increases, from negative to positive, in alkalinity, and a concomitant rise in pH from about 4.3–4.7. Base cations also rose, in contrast to the case with the regional lake survey.

Experimental reduction of acid loading to a whole roofed-over ecosystem in the Norwegian RAIN project (Wright and Hauhs, 1991) resulted in a decline of strong acid anions amounting to 95 μeq l⁻¹. It was compensated by a decline in base cations of 36 μeq l⁻¹ and a rise in alkalinity of 59 μeq l⁻¹. Budgets of input and output showed that long-term acidification of the catchment’s soils has been reversed, so that they are now accumulating base cations. Beier et al. (1995) have recently reviewed modelling studies of recovery in three different experiments.

Reversal of acidification in lake 223 of the Experimental Lakes Area of Ontario has resulted in rapid rises of both alkalinity and pH, due chiefly to
generation of internal alkalinity, similar to the situation near Sudbury (Schindler et al., 1991).

10.3. Biological responses of aquatic ecosystems to lowered acidity

Schindler et al. (1991) reported that recovering lakes near Sudbury have shown rapid increases in the diversity of invertebrate animals, and that reintroduced lake trout have survived and been able to reproduce in several lakes. Recovery of the lakes, and of their biota, has been slow and characterized by the dominance of a few species (Havas et al., 1995). Experimental reversal of acidification in one Ontario lake has yielded mixed results, with some groups recovering rapidly, others more slowly, and a few not at all (Schindler et al., 1991). Obviously, much remains to be learned.

11. Interactions with other environmental stresses

It has recently become apparent that acid deposition interacts with other environmental stresses to damage ecosystems. One interesting example is the penetration of biologically harmful ultraviolet radiation into lakes and streams. Two research groups (Schindler et al., 1996; Yan et al., 1996; see also Gorham, 1996) have shown such penetration to be enhanced not only by depletion of stratospheric ozone but also, through reducing concentrations of dissolved organic matter that absorbs such radiation, by acid deposition and the climatic warming observed in Ontario in recent decades. The effect is significant chiefly in relatively transparent waters (dissolved organic carbon <3.5 mg l⁻¹), which are especially characteristic of large lakes and lakes in arctic, subalpine and alpine regions. An interaction to be expected with future climatic warming and increased drought frequency is the drawdown of peatland water tables, which is likely to result in oxidizing and releasing both anthropogenically derived and other sulfur compounds deposited from the atmosphere and stored in peat (Bayley et al., 1986). Wright and Schindler (1995) have reviewed a variety of interactions between acid deposition and other global changes.

12. Methods employed in studies of acidification and recovery

Several different techniques have been employed to investigate acid deposition and its consequences. The earliest studies of acid deposition and its effects were observations, in the mid-1950s, of correlations between hydrogen ions and sulfate in precipitation (Barrett and Brodin, 1955; Gorham, 1955) and of low pH in lakes on hard igneous rocks that were subject to acid loading from the atmosphere (Gorham, 1955, 1958b; Mackereth, 1957). Similar observations of the biological effects of acid deposition upon aquatic ecosystems were first made in the late 1950s (Dannevig, 1959) and early 1960s (Gorham and Gordon, 1963), and were documented extensively in the 1970s.

Experimental investigations of acid addition to lakes and its chemical and biological effects began in 1976 at the Experimental Lakes Area in Ontario (Schindler, 1980), and were followed by studies during a recovery period in 1984. An experiment on chemical recovery of a whole catchment in Norway (Wright and Hauhs, 1991) also began in 1984. Several experimental additions of nitrogen to forests, designed to simulate the effects of acid deposition, began in the late 1980s and early 1990s (Tietema et al., 1995).

Paleoecological studies of lake acidification in response to acid deposition began in the 1980s (Davis and Berge, 1980); a recent review has been published by Charles et al. (1994). The most detailed account yet of the sequential natural and anthropogenic acidification of a lake was carried out on Lilla Oresjön in Sweden by Renberg (1990).

Modelling exercises, which can be used to project possible future scenarios, began in the late 1970s with Henriksen’s (1980) empirical titration model for acidification based on regional water-chemistry data for more than 700 lakes in Norway. It was followed soon thereafter by the complementary Birkenes model (Christopherson et al., 1982) involving both hydrology and chemistry in accounting for day-to-day variations of stream-water chemistry in acidified regions of southern Norway. Recently, a process-oriented model of intermediate complexity, MAGIC (Beier et al., 1995; Cosby et al., 1985), has been applied with some success to recovery from acidification in the Norwegian RAIN project referred to earlier. It and a number of other models were examined in the summary volume for the 5 year Surface Water Acidification Programme sponsored by the scientific academies of Britain, Sweden and Norway (Mason, 1990), and the subject has been reviewed recently by Rasmussen et al. (1995). According to Schindler (1987), the most effective models for managing aquatic ecosystems have usually been ‘simple and directly grounded in empirical science’.

13. Major international meetings

Research on acid deposition and its effects became an international issue following an influential publication by Svante Öden (1968) in Sweden. He was the first person to suggest that acidifying air pollution transported across international boundaries could
cause a broad range of damage to streams, lakes, upland soils and the biota that they supported, as well as accelerating corrosion of metals and weathering of human artifacts. His concern, expressed also in Dagens Nyheter, a major Stockholm newspaper (Öden, 1967), led to the development of Sweden’s case study for the United Nations Conference on Human Environment (Anonymous, 1971) in Stockholm.


Several major research programs have been of great significance. A large-scale monitoring network for precipitation chemistry was set up in Scandinavia by Egnér and his colleagues (Emanuelsson et al., 1954) and later expanded to cover most of Europe. Following this example, similar networks were set up in 1977 in the United States (National Atmospheric Deposition Program) and Canada (Canadian Network for Sampling Acid Precipitation, later the Canadian Air and Precipitation Monitoring Network). Important roles were played by American scientists at the Hubbard Brook Experimental Forest, beginning in 1964 (Driscoll et al., 1989), and by Norwegian scientists in an 8 year project, the Norwegian Interdisciplinary Research Programme ‘Acid precipitation: effects on forests and fish’, which began in 1972 (Overrein et al., 1980). A massive American program of research was carried out under the National Acid Precipitation Assessment Program, established in 1982 (NAPAP, 1991). Its results were assessed in 1992 in a special feature of five articles in Ecological Applications (Vol. 2, No. 2, pp. 103–130), which ranged from generally favorable to severely critical. Canada’s program of research on the effects of acid deposition on forest and aquatic ecosystems has recently been summarized by Hall et al. (1997) and Jeffries (1997) respectively. The important experimental programs on acidification and recovery in Canada and Norway have already been mentioned, as has the 5 year Surface Water Acidification Programme in the United Kingdom, Norway and Sweden.

14. Policy lessons

The history of four decades of research on acid deposition and its effects has provided several insights about the social dynamics of such research and the policy lessons to be learned from them. Some of these have been described in the above-mentioned series of articles in Ecological Applications. Cowling and Nilsson (1995) focus more specifically upon policy issues. They remark that environmental scientists in this field (as in others) often spend more time and energy in data gathering than in the analysis and interpretation of data. This is especially true of large survey programs and monitoring networks. They discuss reasons of personal and social psychology, as well as budgetary and logistical reasons, for this phenomenon. They also suggest a variety of remedies for such a situation, some social and political, others scientific and intellectual, and yet others involving budgets and logistics.

The same two authors draw ten policy lessons from the history of research on environmental problems such as acid deposition, and note several differences in American and European attitudes and traditions in their approach to decisions on environmental matters. They then proceed to a series of recommendations to enhance decision-making on issues of environmental policy, and conclude with a discussion of ethical considerations.

Another lesson to be learned is that scientists (including this author) often do not take the time to search adequately the earlier research literature, so that they ‘discover’, time and again, what is already known (Gorham, 1981, 1982). Exponential increase in the number of scientific articles published each year makes such a search ever more difficult, despite the availability of computerized techniques.

15. Major advances

A brief chronology of advances, judged by me to be particularly significant in the progress of research on acid deposition and its ecological effects, is presented in Table 1.

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